



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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| (51) International Patent Classification <sup>4</sup> :<br><b>C09J 3/14, A61L 15/06</b>   |  | <b>A1</b>  | (11) International Publication Number: <b>WO 89/ 05334</b><br>(43) International Publication Date: <b>15 June 1989 (15.06.89)</b> |
| (21) International Application Number: <b>PCT/US88/04133</b><br>(22) International Filing Date: <b>21 November 1988 (21.11.88)</b><br>(31) Priority Application Number: <b>126,439</b><br>(32) Priority Date: <b>30 November 1987 (30.11.87)</b><br>(33) Priority Country: <b>US</b><br>(71) Applicant: <b>EASTMAN KODAK COMPANY [US/US]; 343 State Street, Rochester, NY 14650 (US).</b><br>(72) Inventors: <b>FOSTER, Bruce, William ; 617 Ruthlynn, Longview, TX 75601 (US). CLUBB, Clyde, Neal ; 3021 Marlboro Drive, Longview, TX 75605 (US). STUART, Richard, Kingsley, Jr. ; 718 Oakwood Drive, Longview, TX 75604 (US).</b><br>(74) Agent: <b>SAVITSKY, Thomas, R.; 343 State Street, Rochester, NY 14650 (US).</b> |  | (81) Designated States: <b>AT (European patent), BE (European patent), CH (European patent), DE (European patent), FR (European patent), GB (European patent), IT (European patent), JP, LU (European patent), NL (European patent), SE (European patent).</b><br><b>Published</b><br><i>With international search report.</i> |   |
| (54) Title: <b>HOT-MELT ADHESIVE COMPOSITION</b>  |  |  |   |
| (57) Abstract<br><p>The present invention is directed to a hot-melt adhesive containing at least one olefin-based polymer such as a propylene/1-hexene copolymer. The adhesive contains an amount of a low viscosity, substantially crystalline wax sufficient to improve the elastic delamination resistance of the adhesive. The adhesives are particularly useful in disposable diaper construction.</p>   |  |  |   |

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- 1 -

## HOT-MELT ADHESIVE COMPOSITION

### Field of Invention

The present invention concerns hot-melt adhesives (HMAs) containing at least one olefin-based polymer useful as disposable diaper adhesives.

### Background of the Invention

Recent trends in the design of disposable diapers have necessitated the development of more versatile adhesives for use in the diapers. The trend of the industry toward diapers with elastic bands on either the legs or the waist of the diaper have made it necessary to use a stronger more delamination resistant adhesive to hold the elastic in place. Most diaper manufacturers prefer to use one hot-melt adhesive to both construct the diaper (that is to bind the nonwoven top sheet and filler to the polyethylene back sheet) and to hold the elastic on the leg or waist of the diaper. It is well known that polyolefin based HMAs are suitable for the construction of diapers by construction techniques as currently practiced such as multiple fine line, hot-melt spray, hot-melt foam, slot coating operations, and various screen coating methods. However, polyolefin-based HMAs are traditionally not suitable for the bonding of the elastic to the diapers, that is, the elastic delamination resistance is insufficient for such an application. For this reason, adhesives based on styrene rubbers such as S-I-S block copolymers or S-B-S block copolymers are used (see, for example, U.S. Patent 4,526,577).

The use of two HMAs on the same diaper poses some problems for the diaper manufacturer. They must insure the right adhesive is used in the right melt tank and is applied to the correct place on the

- 2 -

diaper. Therefore, an adhesive that is capable of performing both types of bonding functions is highly desirable.

Wax is traditionally added to hot-melt adhesives with the objective of controlling viscosity of the adhesive. That is, it is a diluent and may in some cases lower the cost of the adhesive. For the most part waxes are not known to add to the strength of HMAs but rather are used to control set time or open time or viscosity.

This invention describes a polyolefin-based HMA suitable for use as both the adhesive of construction of disposable diapers and as the adhesive to hold the elastic onto the leg or waistband. Thus a universal diaper adhesive based on polyolefins has been developed. It has been surprisingly found that the addition of a low viscosity, substantially crystalline wax to a polyolefin-based HMA substantially improves the elastic delamination resistance. The formulation of an HMA containing an olefin-based polymer having sufficient elastic delamination resistance to meet the demands of the universal diaper adhesive is a significant advance in the art.

#### 25 Summary of the Invention

The present invention is directed to a hot-melt adhesive composition which has a blend of properties that makes it ideally suited for use with disposable diapers.

30 More specifically, the present invention is directed to a hot-melt adhesive composition having a viscosity of about 3,000 to about 25,000 centipoise at 135°C and a Ring and Ball softening point of about 90°C to about 125°C wherein said adhesive composition contains at least one substantially amorphous

- 3 -

olefin-based polymer with an acid number less than about 0.5, said adhesive composition having blended therein, in an amount sufficient to improve the elastic delamination resistance of the composition, at least one substantially crystalline, low viscosity hydrocarbon wax containing substantially no propylene and having a melt viscosity of from about 3 to about 4,000 centipoise at 150°C and a melting point of from about 90°C to about 125°C.

Preferably, the composition of the present invention is a hot-melt adhesive composition having a viscosity of about 3,000 to about 25,000 centipoise at 135°C and a Ring and Ball softening point of about 90°C to about 125°C, said composition comprising a blend of:

- (a) at least one substantially amorphous olefin-based polymer having an acid number of less than about 0.5,
  - (b) at least one tackifier, and
  - (c) at least one substantially crystalline, low viscosity hydrocarbon wax containing substantially no propylene and having a melt viscosity of from about 3 to about 4,000 centipoise at 150°C and a melting point of from about 90°C to about 125°C,
- the concentration of components (a), (b), and (c) being such that said adhesive composition has an elastic delamination resistance of at least about 4 hours wherein elastic delamination resistance is the length of time a polyethylene to elastic bond can withstand the stress of elastic relaxation at body temperature.

Most preferably, the composition of the present invention is a hot-melt adhesive composition having a viscosity of about 3,000 to about 25,000 centipoise at 135°C and a Ring and Ball softening point of about

- 4 -

90°C to about 125°C, said composition comprising a blend of:

- 5 (i) about 30 to about 70 weight percent of at least one substantially amorphous propylene/C<sub>4</sub> to C<sub>10</sub> higher 1-olefin random copolymer containing from about 20 to about 50 weight percent of higher 1-olefin said polymer having a melt viscosity of from about 2,000 to about 20,000 centipoise at 190°C,
- 10 (ii) about 20 to about 50 weight percent of at least one solid tackifier comprising hydrocarbon resins or polyterpene resins, said solid tackifier having a Ring and Ball softening point of from about 70°C to about 145°C,
- 15 (iii) 0 to about 30 weight percent of at least one liquid tackifier having a viscosity of from about 5 to about 500 centipoise at 95°C and a Ring and Ball softening point of from about 5°C to about 30°C, and
- 20 (iv) from about 1 to about 10 weight percent of at least one substantially crystalline, low viscosity hydrocarbon wax containing substantially no propylene and having a melt viscosity of from about 3 to about 4,000 centipoise at 150°C and a melting point of from about 90°C to
- 25 about 125°C.
- 30

The present invention is also directed to a method for applying the compositions of the invention to a disposable diaper and to articles comprising the composition of the invention in combination with a disposable diaper.

- 5 -

Detailed Description of the Invention

The adhesive compositions of the present invention preferably have a melt viscosity of about 4,000 to about 10,000 centipoise at 135°C. Melt viscosities of the adhesive compositions and the various components thereof can be determined using a Brookfield Model RTV Thermosel viscometer using a Number 27 spindle in accordance with American Society for Testing and Materials (ASTM) Procedure 1824-66.

10 The adhesive compositions of the present invention preferably have a Ring and Ball softening point (RBSP) of about 100°C to about 120°C. RBSP for the adhesive compositions and various components thereof can be determined by use of ASTM  
15 Procedure E-28.

The Gardner color of the adhesive compositions of the present invention preferably is less than about 7, most preferably less than about 3. Gardner color for the adhesive compositions and various  
20 components thereof can be determined by use of ASTM Procedure D-1544.

Bond strength of the adhesives of the present invention is at least about 200 grams based on an adhesive coating weight of 2.1 milligrams (mg) per  
25 inch (0.83 mg/cm), preferably at least 300 grams. At about 300 grams substrate failure occurs. Bond strength can be measured by the controlled destruction of the bonds by an Instron tensile tester at 10 inches (25.4 cm) per minute (min.). More  
30 specifically, nonwoven fabric (e.g., polypropylene or polyester nonwoven fabric commonly used in disposable diaper construction) is bonded by applying a single 2.1 mg per inch (0.83 mg/cm) bead of adhesive on a moving web of polyethylene film at a melt temperature  
35 of 140°C, then applying sufficient nip pressure to bring the two substances in contact. The open time

- 6 -

is about 1 second, and the compression time is less than 0.5 seconds. The nonwoven fabric is then pulled from the polyethylene in a T-peel mode and the average amount of force required to pull apart the two substances is the bond strength.

Tensile strength of the adhesive compositions of the present invention is typically greater than about 10 pounds per square inch (psi) (68.95 kilopascal), and preferably about 20 to about 50 psi (about 137.90 to about 344.74 kilopascal). Tensile strength can be determined by use of ASTM Procedure D-638-72.

The Cone penetration value of the adhesive compositions of the present invention is typically greater than about 10 decimillimeters (dmm), and preferably greater than about 20 dmm. Cone penetration can be measured by use of ASTM Procedure D-1403.

The elastic delamination resistance of the compositions of the present invention is greater than about 4 hours, preferably greater than about 8 hours, more preferably greater than about 24 hours, and most preferably greater than about 100 hours. The elastic delamination resistance is determined by the following procedure:

Elastic, 0.25 inches (0.635 cm) wide and 0.007 (0.01778 cm) inches thick, typically used in disposable diaper construction (can be obtained from Fulflex Company) is expanded to 100% elongation. Test adhesive is applied as a bead at 275°F (135°C) to the elongated elastic which is then immediately brought into contact with polyethylene film and passed through a compression roller. The hot-melt adhesive bead weight is controlled at 10 to 15 mg per linear inch of polyethylene film. The polyethylene film is 1-2 mils thick and is a low density polyethylene blended with linear low density



- 7 -

polyethylene or high density polyethylene. The polyethylene film is pigmented with  $\text{TiO}_2$  and corona treated and is typical of polyethylene film used in disposable diaper manufacture. The bonded material is allowed to relax and age overnight. After aging, specimens 8-inches long are cut. The center 6 inch (15.25 cm) sections of the specimens are elongated to 12 inches (30.48 cm) (100% elongation) and then the polyethylene is clamped in a metal jig with the elastic surface up. The elastic is not clamped. The specimens are then placed in an oven at 100°F (37.78°C) for various times. The time at which a visible, continuous delamination or end release is observed is the elastic delamination resistance.

The adhesive compositions of the present invention contain at least one substantially amorphous olefin-based polymer with an acid number less than about 0.5. The olefin-based polymer is preferably present as an amount of 30 to about 70 weight percent of the adhesive composition, more preferably about 40 to about 65 weight percent. The olefin-based polymer can be a copolymer or a polymer made from more than two monomers. Such polymers are known to be useful in adhesive formulations and many are commercially available. The olefin-based polymers can be prepared using techniques known in the art, for example, by use of the techniques disclosed in U.S. Patents 3,954,697 and 4,259,470, both of which are incorporated herein by reference.

Preferred olefin-based polymers have a melt viscosity of from about 2,000 to about 20,000 centipoise (cp) at 190°C, preferably about 4,000 to about 20,000 cp. Preferred olefin-based polymers are propylene/ $\text{C}_4$  to  $\text{C}_{10}$  higher 1-olefin random copolymers having a higher 1-olefin content of about 20 to about 50 weight percent, preferably about

- 8 -

30 to about 40 weight percent. The preferred higher 1-olefins in the copolymers are 1-hexene and 1-butene with 1-hexene being most preferred. It is preferred that the RBSP for such copolymers is about 100°C to  
5 about 134°C, more preferably about 124°C to about 134°C for a propylene/hexene copolymer and about 100°C to about 120°C for a propylene/butene copolymer.

The low viscosity, substantially crystalline  
10 hydrocarbon wax in the adhesive composition of the present invention is present in an amount sufficient to improve the elastic delamination resistance of the composition. Typically this amount is about 1 to about 10 weight percent of the composition,  
15 preferably about 3 to about 7 weight percent. The hydrocarbon waxes in the adhesive compositions of the present invention are widely available articles of commerce and can be prepared by known techniques. These waxes are preferably polyethylene waxes. Such  
20 polyethylene waxes can be made by direct synthesis or by degrading polyethylene, preferably high density polyethylene (i.e.,  $\geq$  about 0.94 grams(g)/cubic centimeters (cc)) to the desired viscosity. The waxes have a melt viscosity of from about 3 to about  
25 4,000 centipoise, preferably about 3 to about 1,000 centipoise and a melting point of from about 90°C to about 125°C, preferably about 105°C to about 125°C. The waxes also preferably have a penetration hardness at 23°C of about 0.1 dmm to about 10 dmm.  
30 Penetration hardness can be measured by ASTM procedure D-1321-76. Specific waxes useful in the present invention include Epolene C-15 and Epolene N-45, available from Eastman Chemical Products, Inc., Bareco 1000 and Bareco BE<sup>2</sup> 195, available from  
35 Petrolite Corp., and Paraflint H1, available from Moore and Munger, Inc. Preferred is Bareco 1000 and Epolene N-45.

- 9 -

The adhesive compositions of the present invention typically contain one or more tackifiers in an amount of about 20 to about 60 weight percent of the composition. Solid tackifiers are typically present in an amount of from 20 to 50 weight percent, preferably 25 to 40 weight percent. The solid tackifiers can be hydrocarbon resins such as DAC-B hydrocarbon resin prepared according to the process disclosed in U.S. Patent 3,701,760 as well as other hydrocarbon resins, polyterpenes, and the like. One such hydrocarbon tackifying resin is a hydrocarbon resin having a softening point of about 100°C and available commercially as Eastotac H-100 from Eastman Chemical Products, Inc. Other hydrocarbon tackifying resins can be prepared by the polymerization of monomers consisting primarily of olefins and diolefins and include, for example, the residual by-product monomers resulting from the manufacture of isoprene. These hydrocarbon tackifying resins typically exhibit a RBSP of from about 70°C to about 145°C; an acid number of from about 0-2, a saponification value of less than about 1; and an iodine value of from about 30 to 100. Examples of such commercially available resins based on a C<sub>5</sub>-olefin fraction of this type are "Wingtack" 95 and "Wingtack" 115 tackifying resins sold by Goodyear Tire and Rubber Company, the Sta-Tac and Betaprene A or H resins sold by Reichhold Chemical Corporation, and Escorez resins sold by Exxon Chemical Co.

Also, other suitable solid tackifier resins are the terpene polymers such as the polymeric, resinous materials obtained by polymerization and/or copolymerization of terpene hydrocarbons such as the alicyclic, monocyclic, and bicyclic monoterpenes and their mixtures, including alloocimene, carene, isomerized pinene, pinene, dipentene, terpinene,

- 10 -

terpinolene, limonene, terpine, a terpene cut or fraction, and various other terpenes. Particularly useful starting materials are terpene mixtures containing at least 20 percent beta-pinene and/or  
5 limonene or dipentene (racemic limonene), and the "sulfate terpine" obtained as a by-product in the sulfate pulping process. Commercially available resins of the terpene type include the Zonarez terpene B-Series and 7000 Series resins from Arizona  
10 Chemical Corp. and Nirez resins from Reichhold Chemical Corp. The typical properties reported for the Zonarez terpene resins include RBSPs of about 55° to 125°C (ASTM E-28-67), color of 2 to 3 (Gardner 1963, 50% in heptane), acid number of less than 1  
15 (ASTM D465-59), saponification number of less than 1 (ASTM D464-59) and specific gravity at 25°C of 0.96 to 0.99 (ASTM D1963-61).

The liquid tackifiers are typically present in an amount of from 0 to about 30 weight percent of the  
20 adhesive composition, preferably 15 to 25 weight percent. The liquid tackifiers are commercial items and/or can be prepared by techniques known in the art, for example, by the techniques described in U.S. Patent 3,872,064, incorporated herein by reference.  
25 The liquid tackifiers are preferably liquid hydrocarbon resins such as synthetic polyterpene or other petroleum hydrocarbon resins. Specific examples include "Wingtack" 10 from Goodyear Tire and Rubber Company and "Escorez" 2520 (also known as  
30 ECR-140) from Exxon Chemical Co. The liquid tackifiers have a melt viscosity of about 10,000 to about 50,000 cp at 23°C, preferably about 20,000 to about 40,000; a RBSP of about 5 to about 30, preferably about 10 to about 20, and a glass  
35 transition temperature (T<sub>g</sub>) of about -10°C to about -30°C.

- 11 -

The adhesive compositions of this invention are prepared by blending together the adhesive components in the melt at a temperature of about 160°C to about 200°C until a homogeneous mixture is obtained.

5 Various methods of blending materials of this type are known to the art and any method that produces a homogeneous mixture is satisfactory. These components blend easily in the melt and a heated vessel equipped with a stirrer is all that is  
10 required. For example, a Cowles stirrer provides effective mixing for preparing these compositions. Solvents such as hexane, heptane, mineral spirits, xylene, toluene, benzene, chlorinated hydrocarbons, etc., are not needed to prepare the compositions of  
15 this invention; however, they can be used if desired.

In addition to the hereinabove described adhesive components, it is desirable for the adhesive compositions to contain about 0.1 percent to about  
20 1.5 percent by weight, preferably about 0.25 percent to 1.0 percent by weight, of one or more antioxidants. Antioxidants that are effective include, for example, tris(di-t-butyl-p-hydroxybenzyl)-trimethylbenzene (Ionox 330), alkylated  
25 bisphenol (Naugawhite), zinc dibutyl dithiocarbamate (Butyl Zimate), and 4,4'-methylene bis(2,6-di-tert-butylphenol)(Ethyl 702), tetrakis[methylene(3,5-di-tert-butyl-4-hydroxyhydrocinnamate(methane)]  
(Irganox 1010), lauryl stearyl thiodipropionate  
30 (Cyanox 1212), and dilauryl 3,3'-thiodipropionate (Cyanox LTDP) and 2,6-di-tert-butyl-p-cresol (BHT) and the like.

Additional additives such as nucleating agents, pigments, colorants, fillers, solvents, and the like  
35 can also be added to the adhesive compositions of the present invention.

- 12 -

The adhesive compositions of the present invention can be made into any physical form typically used in the art. When formed into slats, the compositions typically are coated with a wax which may or may not be a wax within the scope of the invention.

In the method of the present invention, the adhesive composition is applied to a disposable diaper or portion thereof using techniques known in the art. For example, the adhesive composition can be applied by multiple fine line, hot-melt spray, hot-melt foam, slot coating operations, and various screen coating methods. The amount of adhesive composition applied to a diaper is that amount sufficient to result in bonds that will withstand typical storage and end use conditions. It is contemplated that the adhesive compositions of the present invention are useful in applications other than diapers, for example, in sanitary napkins and bed pad construction.

In the adhesive compositions of the present invention, as appreciated by one skilled in the art, the particular proportions of components necessary to achieve specific desired properties will vary depending on the nature of the particular components.

This invention can be further illustrated by the following examples thereof, although it will be understood that these examples are included merely for purposes of illustration and are not intended to limit the scope of the invention.

#### Examples 1-2

An adhesive composition was made in accordance with the present invention containing 5 weight % of a hydrocarbon wax as described herein (Example 2). For comparison, an adhesive composition outside the scope of the invention, i.e., not containing a hydrocarbon

- 13 -

wax, was also prepared (Example 1). The two compositions were tested for their respective elastic delamination resistance (EDL). As shown in Table 1, the EDL of Example 1 was only four hours, whereas the  
5 EDL of Example 2 was greater than 100 hours.

- 14 -

TABLE 1 - ADHESIVE BLENDS

| EXAMPLE NUMBER                  | 1              | 2                              |
|---------------------------------|----------------|--------------------------------|
| COMPOSITION (Weight %)          |                |                                |
| <sup>1</sup> APH 5              | 26             | 26                             |
| <sup>2</sup> APH 3              | 17             | 22                             |
| <sup>3</sup> Eastotac H-130W    | 36.5           | 26.5                           |
| <sup>4</sup> Escorez 2520       | 20             | 20                             |
| <sup>5</sup> Bareco 1000        | -              | 5.0                            |
| <sup>6</sup> Irganox 1010       | 0.5            | 0.5                            |
| BLEND PROPERTIES                |                |                                |
| VISCOSITY @ 135°C, cp           | 9600           | 6600                           |
| RBSP, °C                        | 113            | 108                            |
| CONE PENETRATION, dmm           | 18             | 14                             |
| TENSILE STRENGTH, psi (KPa)     | 15<br>(103.42) | 30<br>(206.84)                 |
| ELASTIC DELAMINATION RESISTANCE |                |                                |
| Time to failure, hrs            | 4              | >100                           |
| BOND STRENGTH, grams            | 200            | 300 (sub-<br>strate<br>failed) |

- <sup>1</sup>APH 5 = amorphous propylene/1-hexene copolymer having a viscosity of 17,000 cp at 190°C, 23 weight % 1-hexene content, and a RBSP of 138°C.
- <sup>2</sup>APH 3 = amorphous propylene/1-hexene copolymer having a viscosity of 3,000 cp at 190°C, 55 weight % 1-hexene content, and a RBSP of 114°C.
- <sup>3</sup>Eastotac H-130W = solid tackifier, petroleum hydrocarbon resin, RBSP of 130°C melt viscosity at 190°C of 1,000 cp.
- <sup>4</sup>Escorez 2520 = liquid tackifier, RBSP of 20°C, T<sub>g</sub> of -20°C, melt viscosity of 40,000 cp at 23°C.
- <sup>5</sup>Bareco 1000 = high density, low viscosity, crystalline polyethylene wax, melting point of 113°C, penetration hardness of 1 dmm, density of 0.96 g/cc, melt viscosity of 11 cp at 150°C.
- <sup>6</sup>Irganox 1010 = antioxidant



- 15 -

Examples 3-8

Adhesive blends having a relatively high proportion of propylene/1-hexene copolymer (i.e. 47%) were prepared in accordance with the present invention. The RBSPs of the blends were varied primarily by adjusting the % hexene content of the copolymer. Variations in the elastic delamination resistance (EDL) were observed, however, in all cases the EDLs were greater than four hours as shown in Table 2.

- 16 -

TABLE 2 - ADHESIVE BLENDS

| EXAMPLE NUMBER                         | 3    | 4     | 5     | 6     | 7    | 8    |
|--|------|-------|-------|-------|------|------|
| <b>BLEND COMPOSITION (Wt %)</b>        |      |       |       |       |      |      |
| <sup>1</sup> APH                       | 47.0 | 47.0  | 47.0  | 47.0  | 47.0 | 47.0 |
| <sup>2</sup> Escorez 2520              | 19.5 | 19.5  | 19.5  | 19.5  | 19.5 | 19.5 |
| <sup>3</sup> Eastotac H-130W           | 26.0 | 26.0  | 26.0  | 26.0  | 26.0 | 26.0 |
| <sup>4</sup> Bareco 1000               | 3.0  | 3.0   | 3.0   | 3.0   | 3.0  | 3.0  |
| <sup>5</sup> Epolene C-15              | 4.0  | 4.0   | 4.0   | 4.0   | 4.0  | 4.0  |
| <sup>6</sup> Irganox 1010              | 0.5  | 0.5   | 0.5   | 0.5   | 0.5  | 0.5  |
| <b>PHYSICAL PROPERTIES OF APH</b>      |      |       |       |       |      |      |
| Viscosity @<br>375°F(190.56°C), cp     | 5475 | 8325  | 7650  | 9550  | 7000 | 5300 |
| RBSP, deg C                            | 132  | 138   | 135   | 132   | 124  | 126  |
| % Hexene                               | 36   | 33    | 35    | 36    | 40   | 40   |
| <b>PHYSICAL PROPERTIES OF BLEND</b>    |      |       |       |       |      |      |
| Viscosity @ 135°C, cp                  | 8500 | 22750 | 29000 | 19000 | 7750 | 6440 |
| RBSP, °C                               | 111  | 120.5 | 116.5 | 112   | 107  | 107  |
| Cone Penetration, dmm                  | 25   | 23.5  | 24    | 22    | 23   | 26   |
| <b>ELASTIC DELAMINATION RESISTANCE</b> |      |       |       |       |      |      |
| Time to failure, hrs                   | >47  | >47   | >4    | >4    | >4   | >8   |

<sup>1</sup>APH = amorphous propylene/1-hexene copolymer

<sup>2</sup>Escorez 2520 = liquid tackifier, RBSP of 20°C, T<sub>g</sub> of -20°C, melt viscosity at 23°C of 40,000 cp.

<sup>3</sup>Eastotac H-130W = solid tackifier, petroleum hydrocarbon resin, RBSP of 130°C, melt viscosity at 190°C of 1,000 cp.

<sup>4</sup>Bareco 1000 = high density, low viscosity, crystalline polyethylene wax, melting point of 113°C, penetration hardness of 1 dmm, density of 0.96 g/cc, melt viscosity of 11 cp at 150°C.

<sup>5</sup>Epolene C-15 = lower density, low viscosity, crystalline polyethylene wax, melting point of 102°C, penetration hardness of 4 dmm, density of 0.91 g/cc, melt viscosity of 3,900 cp at 150°C.

<sup>6</sup>Irganox 1010 = antioxidant

- 17 -

Examples 9-16

Adhesive blends were prepared in accordance with the present invention. The compositions and properties of these blends are shown in Table 3. Example 14 is  
5 a comparative example without a wax.

TABLE 3 - ADHESIVE BLENDS

| EXAMPLE NUMBER              | COMPOSITION (Wt. %)                      | 9    | 10   | 11   | 12   | 13   | 14   | 15   | 16   |
|-----------------------------|--|------|------|------|------|------|------|------|------|
| 1APH                        |  | 48   | -    | 48   | 48   | 48   | 50.5 | 50   | 45.7 |
| 2APB                        |  | -    | 54.5 | -    | -    | -    | -    | -    | -    |
| 3Eastotac H-130W            |  | 26.5 | 20   | 26.5 | 26.5 | 26.5 | 27.9 | 17.6 | 25.2 |
| 4Escorez 2520               |  | 20   | 20   | 20   | 20   | 20   | 21.1 | 20.8 | 19   |
| 5Bareco 1000                |  | 5    | -    | -    | -    | -    | -    | -    | -    |
| 6Epolene N-45               |  | -    | 5    | -    | -    | -    | -    | 1    | 10   |
| 7Epolene C-15               |  | -    | -    | 5    | -    | -    | -    | -    | -    |
| 8Bareco Be <sup>2</sup> 195 |  | -    | -    | -    | 5    | -    | -    | -    | -    |
| 9Paraflint H-1              |  | -    | -    | -    | -    | 5    | -    | -    | -    |
| 10Irganox 1010              |  | 0.5  | 0.5  | 0.5  | 0.5  | 0.5  | 0.5  | 0.5  | 0.5  |
| 1APH                        | = amorphous propylene/1-hexene copolymer |      |      |      |      |      |      |      |      |
| 2APB                        | = amorphous propylene/1-butene copolymer |      |      |      |      |      |      |      |      |
| 3Eastotac H-130W            | = solid tackifier                        |      |      |      |      |      |      |      |      |
| 4Escorez 2520               | = liquid tackifier                       |      |      |      |      |      |      |      |      |
| 5Bareco 1000                | = wax                                    |      |      |      |      |      |      |      |      |
| 6Epolene N-45               | = wax                                    |      |      |      |      |      |      |      |      |
| 7Epolene C-15               | = wax                                    |      |      |      |      |      |      |      |      |
| 8Bareco Be <sup>2</sup> 195 | = wax                                    |      |      |      |      |      |      |      |      |
| 9Paraflint H-1              | = wax                                    |      |      |      |      |      |      |      |      |
| 10Irganox 1010              | = antioxidant                            |      |      |      |      |      |      |      |      |

- 18 -

TABLE 3- ADHESIVE BLENDS (Continued)

| EXAMPLE NUMBER                     | 9        | 10       | 11       | 12                | 13              | 14   | 15       | 16       |
|------------------------------------|----------|----------|----------|-------------------|-----------------|------|----------|----------|
| PHYSICAL PROPERTIES OF APH AND APB |          |          |          |                   |                 |      |          |          |
| Viscosity @                        |          |          |          |                   |                 |      |          |          |
| 375°F(190.56°C), cp                | 5475     | 10000    | 5475     | 5475              | 5475            | 5475 | 5475     | 5475     |
| RBSP, °C                           | 132      | 110      | 132      | 132               | 132             | 132  | 132      | 132      |
| % Hexene                           | 36       | 0        | 36       | 36                | 36              | 36   | 36       | 36       |
| % Butene                           | 0        | 40       | 0        | 0                 | 0               | 0    | 0        | 0        |
| PHYSICAL PROPERTIES OF WAX         |          |          |          |                   |                 |      |          |          |
| Melting point, °C                  | 113      | 123      | 102      | 91                | 13              | -    | 123      | 123      |
| Density, g/cc                      | 0.96     | 0.95     | 0.91     | 0.93              | 0.94            | -    | 0.95     | 0.95     |
| Viscosity @ 150°C, cp              | 11       | 500      | 3900     | 3                 | 3               | -    | 500      | 500      |
| Wax type                           | ethylene | ethylene | ethylene | micro-crystalline | Fischer-Tropsch | -    | ethylene | ethylene |
| Penetration Hardness, dmm          | 0.5      | 0.1      | 4        | 7                 | .2              | -    | 0.1      | 0.1      |
| BLEND PROPERTIES                   |          |          |          |                   |                 |      |          |          |
| VISCOSITY @ 135°C, cp              | 8000     | 10500    | 8000     | 4880              | 4880            | 6880 | 7100     | 4200     |
| RBSP, °C                           | 117      | 117      | 117      | 111               | 103             | 113  | 115      | 117      |
| CONE PENETRATION, dmm              | 17       | 18       | 39       | 10                | 18              | 42   | 33       | 14       |
| ELASTIC DELAMINATION RESISTANCE    |          |          |          |                   |                 |      |          |          |
| Time to failure, hrs               | 24       | 24       | 24       | 24                | 24              | <1   | 24       | >72      |

- 20 -

CLAIMS

We claim:

1. A hot-melt adhesive composition having a melt viscosity of 3,000 to 25,000 centipoise at 135°C and a Ring and Ball softening point of 90°C to 125°C wherein said adhesive composition contains at least one substantially amorphous olefin-based polymer with an acid number less than 0.5, said adhesive composition having blended therein, in an amount sufficient to improve the elastic delamination resistance of the composition, at least one substantially crystalline, low viscosity hydrocarbon wax containing substantially no propylene and having a melt viscosity of from 3 to 4,000 centipoise at 150°C and a melting point of from 90°C to 125°C.
2. The composition of Claim 1 wherein said elastic delamination resistance is at least 4 hours.
3. The composition of Claim 1 wherein said elastic delamination resistance is at least 8 hours.
4. The composition of Claim 1 wherein said elastic delamination resistance is at least 24 hours.
5. The composition of Claim 1 which has a cone penetration value as measured by ASTM Procedure D-1403 of at least 10 dmm, a molten Gardner color as measured by ASTM Procedure D-1544 of less than 7, a tensile strength as measured by ASTM Procedure D-638-72 of at least (68.95 KPa), and a bond strength of at least

- 21 -

200 grams per 2.1 mg per inch (0.83 mg/cm)  
coating weight.

- 5 6. The composition of Claim 5 wherein said elastic delamination resistance is at least 8 hours, said cone penetration value is at least 20 dmm, said molten Gardner color is less than 4, said tensile strength is 20 to 50 psi (137.90 to 344.74 KPa), and said bond strength is at least  
10 300 grams per 2.1 mg per inch (0.83 mg/cm) coating weight.
7. The composition of Claim 1 additionally containing at least one tackifier, at least one antioxidant, at least one additional additive or a mixture thereof.
- 15 8. The composition of Claim 1 wherein said adhesive composition has a viscosity of 4,000 to 8,000 centipoise at 135°C and a Ring and Ball softening point of 100°C to 120°C.
- 20 9. The composition of Claim 1 wherein said hydrocarbon wax is a polyethylene wax having a melt viscosity of from 3 to 4,000 centipoise at 150°C and a penetration hardness at 23°C of 0.1 to 10, and a melting point of 105°C to 125°C.
- 25 10. Hot-melt adhesive composition having a melt viscosity of 3,000 to 25,000 centipoise at 135°C and a Ring and Ball softening point of 90°C to 125°C, said composition comprising a blend of:
  - (a) at least one substantially amorphous olefin-based polymer having an acid number  
30 of less than 0.5,

- 22 -

(b) at least one tackifier, and

(c) at least one substantially crystalline, low viscosity hydrocarbon wax containing substantially no propylene and having a melt viscosity of from 3 to 4,000 centipoise at 150°C and a melting point of from 90°C to 125°C.

the concentrations of components (a), (b), and (c) being such that said adhesive composition has an elastic delamination resistance of at least 4 hours wherein elastic delamination resistance is the length of time a polyethylene to elastic bond can withstand the stress of elastic relaxation at body temperature.

11. The adhesive composition of Claim 10 wherein said adhesive composition has a viscosity of 4,000 to 10,000 centipoise at 135°C and a Ring and Ball softening point of 100°C to 120°C.

12. The composition of Claim 10 wherein said hydrocarbon wax is a polyethylene wax having a melt viscosity of from 3 to 1,000 centipoise at 150°C, a melting point of from 105°C to 125°C, and a penetration hardness of 0.1 to 10.

13. The adhesive composition of Claim 10 wherein component (a) is present in an amount of 30 to 70 weight percent, component (b) is present in an amount of 20 to 60 weight percent, and component (c) is present in an amount of from 1 to 10 weight percent.



- 23 -

14. The adhesive composition of Claim 10 wherein said elastic delamination resistance is at least eight hours.
- 5 15. The adhesive compositions of Claim 10 which has a cone penetration as measured by ASTM Procedure D-1403 of at least 10 dmm, a molten Gardner color as measured by ASTM Procedure D-1544 of less than 7, a tensile strength as measured by ASTM Procedure D-638-72 of at least 10 psi (68.95 KPa), a bond strength of at least 200 grams per 2.1 mg per inch (0.83 mg/cm) coating weight, and wherein said elastic delamination resistance is at least 24 hours.
- 10 16. The adhesive composition of Claim 15 wherein said cone penetration value is at least 20 dmm, said molten Gardner color is less than 4, said tensile strength is 20 to 50 psi (137.90 to 344.74 KPa), said bond strength is at least 300 grams per 2.1 mg per inch (0.83 mg/cm) coating weight, and said elastic delamination resistance is at least about 100 hours.
- 15 17. The composition of Claim 10 additionally containing at least one antioxidant, at least one additional additive or a mixture thereof.
- 20 18. A hot-melt adhesive composition having a melt viscosity of 3,000 to 25,000 centipoise at 135°C and a Ring and Ball softening point of 90°C to 125°C, said composition comprising a blend of:
- 25 (i) 30 to 70 weight percent of at least one substantially amorphous propylene/C<sub>4</sub> to
- 30

- 24 -

- 5                   C<sub>10</sub> higher 1-olefin random copolymer  
                  containing from 20 to 50 weight percent  
                  of higher 1-olefin said copolymer having  
                  a melt viscosity of from 2,000 to  
                  20,000 centipoise at 190°C,
- 10                   (ii) 20 to 50 weight percent of at least one  
                  solid tackifier comprising hydrocarbon  
                  resins or polyterpene resins said  
                  tackifier having a softening point of  
                  from 70°C to 145°C.
- 15                   (iii) 0 to 30 weight percent of at least one  
                  liquid tackifier having a viscosity of  
                  from 10,000 to 50,000 centipoise at 23°C  
                  and a Ring and Ball softening point of  
                  from 5°C to 30°C, and
- 20                   (iv) from 1 to 10 weight percent of at least  
                  one high density, substantially  
                  crystalline, low viscosity  
                  hydrocarbon wax containing substantially  
                  no propylene and having a melt viscosity  
                  of from 3 to 4,000 centipoise at 150°C  
                  and a melting point of from 90°C to  
                  125°C.
- 25                   19. The adhesive composition of Claim 18 which has a  
                  cone penetration value as measured by ASTM  
                  Procedure D-1403 of at least 10 dmm, a molten  
                  Gardner color as measured by ASTM  
                  Procedure D-1544 of less than 7, a tensile  
30                   strength as measured by ASTM Procedure D-638-72  
                  of at least 10 psi (68.95 KPa), a bond strength  
                  of at least 200 grams per 2.1 mg per inch (0.83  
                  mg/cm) coating weight, and wherein said elastic

- 25 -

delamination resistance is at least 4 hours.

20. The adhesive composition of Claim 18 wherein said cone penetration value is at least 20 dmm, said molten Gardner color is less than four, said tensile strength is 20 to 50 psi (137.90 to 344.74 KPa), said bond strength is at least 300 grams per 2.1/mg per inch (0.83 mg/cm) coating weight, and said elastic delamination resistance is at least 8 hours.
21. The composition of Claim 20 wherein said delamination resistance is at least 24 hours.
22. The composition of Claim 18 wherein component (i) comprises: 40 to 65 weight percent of at least one substantially amorphous propylene/C<sub>4</sub> to C<sub>10</sub> higher 1-olefin random copolymer containing from 30 to 40 weight percent at higher 1-olefin said copolymer having a melt viscosity of from 4,000 to 20,000 centipoise at 190°C and having a Ring and Ball softening point of 100°C to 134°C.
- Component (ii) comprises: 25 to 40 weight percent of at least one solid tackifier, comprising hydrocarbon resins or polyterpene resins said solid tackifier having a softening point of from 120°C to 145°C,
- Component (iii) comprises: from 15 to 25 weight percent of at least one liquid tackifier having a viscosity of from 20,000 to 40,000 centipoise at 23°C, a Ring and Ball softening point of from about 10°C to about 20°C, and a Tg of about -10°C to about -30°C, and

- 26 -

5           Component (iv) comprises: from about 3 to about 7 weight percent of at least one substantially crystalline, low viscosity polyethylene wax having a melt viscosity of from about 3 to about 1,000 centipoise at 150°C and a melting point of from about 105°C to about 125°C

23.   The adhesive composition of Claim 18 wherein said higher 1-olefin is 1-hexene or 1-butene.
- 10   24.   The adhesive composition of Claim 22 wherein said higher 1-olefin is 1-hexene or 1-butene.
25.   The adhesive composition of Claim 18 wherein said solid tackifier is a C<sub>5</sub> hydrocarbon resin or polyterpene resin.
- 15   26.   The adhesive composition of Claim 24 wherein said solid tackifier is a C<sub>5</sub> hydrocarbon resin or polyterpene resin.
27.   The adhesive composition of Claim 18 wherein said liquid tackifier is a synthetic polyterpene.
- 20   28.   The adhesive composition of Claim 26 wherein said liquid tackifier is a syntehtic polyterpene.
29.   The adhesive composition of Claim 18 additionally comprising an antioxidant, an additional additive or a mixture thereof.
- 25   30.   The adhesive composition of Claim 28 additionally comprising an antioxidant, an additional additive or a mixture thereof.

- 27 -

31. A method comprising applying the adhesive composition of Claim 1 to a disposable diaper or portion thereof.
32. A method comprising applying the adhesive composition of Claim 10 to a disposable diaper or portion thereof.
33. A method comprising applying the adhesive composition of Claim 18 to a disposable diaper or portion thereof.
34. An article comprising the adhesive composition of Claim 1 in combination with a disposable diaper.
35. An article comprising the adhesive composition of Claim 10 in combination with a disposable diaper.
36. An article comprising the adhesive composition of Claim 18 in combination with a disposable diaper.

# INTERNATIONAL SEARCH REPORT

International Application No PCT/US 88/04133

| <b>I. CLASSIFICATION OF SUBJECT MATTER</b> (If several classification symbols apply, indicate all) <sup>6</sup><br>According to International Patent Classification (IPC) or to both National Classification and IPC<br>IPC4: C 09 J 3/14, A 61 L 15/06  |  |                                     |   |  |  |   |  |             |   |  |      |   |   |                 |   |  |                    |
|--|--|-------------------------------------|---|--|--|---|--|-------------|---|--|------|---|---|-----------------|---|--|--------------------|
| <b>II. FIELDS SEARCHED</b><br><div style="text-align: center; border-top: 1px solid black; border-bottom: 1px solid black; margin: 5px 0;">Minimum Documentation Searched <sup>7</sup></div> <table style="width: 100%; border-collapse: collapse;"> <tr> <th style="width: 30%; border-bottom: 1px solid black;">Classification System</th> <th style="width: 70%; border-bottom: 1px solid black;">Classification Symbols</th> </tr> <tr> <td style="padding: 5px; vertical-align: top;">IPC4</td> <td style="padding: 5px; vertical-align: top;">C 08 L, C 09 J</td> </tr> </table> <div style="text-align: center; border-top: 1px solid black; border-bottom: 1px solid black; margin: 5px 0;">Documentation Searched other than Minimum Documentation to the extent that such Documents are included in the Fields Searched <sup>8</sup></div>   |  |                                     | Classification System   | Classification Symbols   | IPC4   | C 08 L, C 09 J  |  |             |   |  |      |   |   |                 |   |  |                    |
| Classification System  | Classification Symbols   |                                     |   |  |  |   |  |             |   |  |      |   |   |                 |   |  |                    |
| IPC4   | C 08 L, C 09 J   |                                     |   |  |  |   |  |             |   |  |      |   |   |                 |   |  |                    |
| <b>III. DOCUMENTS CONSIDERED TO BE RELEVANT <sup>9</sup></b> <table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th style="width: 10%; border-bottom: 1px solid black;">Category <sup>9</sup></th> <th style="width: 60%; border-bottom: 1px solid black;">Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup></th> <th style="width: 30%; border-bottom: 1px solid black;">Relevant to Claim No. <sup>12</sup></th> </tr> </thead> <tbody> <tr> <td style="text-align: center; vertical-align: top; padding: 5px;">X</td> <td style="padding: 5px;">GB, A, 1302283 (BRUNO ROMANIN) 4 January 1973, see page 4, lines 31-37, 64-75, 89, 92-93 and example 1<br/>--</td> <td style="text-align: center; vertical-align: top; padding: 5px;">1-17, 31-36</td> </tr> <tr> <td style="text-align: center; vertical-align: top; padding: 5px;">X</td> <td style="padding: 5px;">US, A, 4471086 (BRUCE W. FOSTER) 11 September 1984, see column 2, line 8 - line 11; column 2, line 52 - line 57; column 3, line 29 - line 40; column 4, line 18 - line 22; claim 1<br/>--</td> <td style="text-align: center; vertical-align: top; padding: 5px;">1-17</td> </tr> <tr> <td style="text-align: center; vertical-align: top; padding: 5px;">X</td> <td style="padding: 5px;">Derwent's abstract No. 31 693 K/13, SU 929 677, publ. week 8313<br/>--</td> <td style="text-align: center; vertical-align: top; padding: 5px;">1, 7, 9-10, 12-</td> </tr> <tr> <td style="text-align: center; vertical-align: top; padding: 5px;">A</td> <td style="padding: 5px;">DE, A, 1960467 (CONTINENTAL CAN COMPANY, INC) 10 September 1970, see claim 1<br/>--</td> <td style="text-align: center; vertical-align: top; padding: 5px;">1-8, 10, 11, 13-17</td> </tr> </tbody> </table> |  |                                     | Category <sup>9</sup>   | Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup> | Relevant to Claim No. <sup>12</sup>                                | X   | GB, A, 1302283 (BRUNO ROMANIN) 4 January 1973, see page 4, lines 31-37, 64-75, 89, 92-93 and example 1<br>-- | 1-17, 31-36 | X | US, A, 4471086 (BRUCE W. FOSTER) 11 September 1984, see column 2, line 8 - line 11; column 2, line 52 - line 57; column 3, line 29 - line 40; column 4, line 18 - line 22; claim 1<br>-- | 1-17 | X | Derwent's abstract No. 31 693 K/13, SU 929 677, publ. week 8313<br>-- | 1, 7, 9-10, 12- | A | DE, A, 1960467 (CONTINENTAL CAN COMPANY, INC) 10 September 1970, see claim 1<br>-- | 1-8, 10, 11, 13-17 |
| Category <sup>9</sup>  | Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup>   | Relevant to Claim No. <sup>12</sup> |   |  |  |   |  |             |   |  |      |   |   |                 |   |  |                    |
| X  | GB, A, 1302283 (BRUNO ROMANIN) 4 January 1973, see page 4, lines 31-37, 64-75, 89, 92-93 and example 1<br>--   | 1-17, 31-36                         |   |  |  |   |  |             |   |  |      |   |   |                 |   |  |                    |
| X  | US, A, 4471086 (BRUCE W. FOSTER) 11 September 1984, see column 2, line 8 - line 11; column 2, line 52 - line 57; column 3, line 29 - line 40; column 4, line 18 - line 22; claim 1<br>-- | 1-17                                |   |  |  |   |  |             |   |  |      |   |   |                 |   |  |                    |
| X  | Derwent's abstract No. 31 693 K/13, SU 929 677, publ. week 8313<br>--  | 1, 7, 9-10, 12-                     |   |  |  |   |  |             |   |  |      |   |   |                 |   |  |                    |
| A  | DE, A, 1960467 (CONTINENTAL CAN COMPANY, INC) 10 September 1970, see claim 1<br>--   | 1-8, 10, 11, 13-17                  |   |  |  |   |  |             |   |  |      |   |   |                 |   |  |                    |
| <div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> <p><sup>10</sup> Special categories of cited documents:</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 45%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"&amp;" document member of the same patent family</p> </div> </div>   |  |                                     |   |  |  |   |  |             |   |  |      |   |   |                 |   |  |                    |
| <b>IV. CERTIFICATION</b> <table style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 50%; border-bottom: 1px solid black; padding: 5px;">           Date of the Actual Completion of the International Search<br/> <b>13th March 1989</b> </td> <td style="width: 50%; border-bottom: 1px solid black; padding: 5px;">           Date of Mailing of this International Search Report<br/> <b>23 MAR 1989</b> </td> </tr> <tr> <td style="border-bottom: 1px solid black; padding: 5px;">           International Searching Authority<br/> <b>EUROPEAN PATENT OFFICE</b> </td> <td style="border-bottom: 1px solid black; padding: 5px;">           Signature of Authorized Officer<br/> <b>P.C.G. VAN DER PUTTEN</b> </td> </tr> </table>   |  |                                     | Date of the Actual Completion of the International Search<br><b>13th March 1989</b> | Date of Mailing of this International Search Report<br><b>23 MAR 1989</b>                                      | International Searching Authority<br><b>EUROPEAN PATENT OFFICE</b> | Signature of Authorized Officer<br><b>P.C.G. VAN DER PUTTEN</b> |  |             |   |  |      |   |   |                 |   |  |                    |
| Date of the Actual Completion of the International Search<br><b>13th March 1989</b>  | Date of Mailing of this International Search Report<br><b>23 MAR 1989</b>  |                                     |   |  |  |   |  |             |   |  |      |   |   |                 |   |  |                    |
| International Searching Authority<br><b>EUROPEAN PATENT OFFICE</b>   | Signature of Authorized Officer<br><b>P.C.G. VAN DER PUTTEN</b>  |                                     |   |  |  |   |  |             |   |  |      |   |   |                 |   |  |                    |

## III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)

| Category * | Citation of Document, with indication, where appropriate, of the relevant passages   | Relevant to Claim No |
|------------|--|----------------------|
| X          | DE, A1, 2613587 (EASTMAN KODAK CO)<br>14 October 1976, see page 4, line 29 -<br>page 5, line 9; page 11, line 19 -<br>line 27<br>--  | 18-30                |
| A          | US, A, 4299745 (DARRYL A. GODFREY)<br>10 November 1981, see column 1,<br>line 35; claim 1<br>--  | 31-36                |
| X          | Dialog Information Services, File 351,<br>World Patent Index 81-89, Dialog accession<br>no. 3780842, Mitsui Dupont Polyc:<br>"Hot-melt adhesive compsn. esp. for<br>smooth materials comprises ethylene-<br>alpha olefin copolymer, ethylene-vinyl<br>ester copolymer, tackifier resin and wax"<br>JP 60051768, A, 850323, 8518 (Basic)<br>--<br>----- | 18-30                |

**ANNEX TO THE INTERNATIONAL SEARCH REPORT  
ON INTERNATIONAL PATENT APPLICATION NO.**

**PCT/US 88/04133**

**SA 25735**

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.  
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| Patent document<br>cited in search report | Publication<br>date | Patent family<br>member(s) | Publication<br>date |
|---|---------------------|----------------------------|---------------------|
| GB-A- 1302283                             | 04/01/73            | NONE                       |                     |
| US-A- 4471086                             | 11/09/84            | NONE                       |                     |
| DE-A- 1960467                             | 10/09/70            | NL-A- 7001612              | 18/08/70            |
|   |                     | FR-A- 2030440              | 13/11/70            |
|   |                     | GB-A- 1245187              | 08/09/71            |
|   |                     | CH-A- 506606               | 30/04/71            |
|   |                     | SE-A-C- 377133             | 23/06/75            |
| DE-A1- 2613587                            | 14/10/76            | US-A- 3954697              | 04/05/76            |
|   |                     | FR-A-B- 2306248            | 29/10/76            |
|   |                     | US-A- 4072812              | 07/02/78            |
|   |                     | GB-A- 1529895              | 25/10/78            |
|   |                     | JP-A- 51122136             | 26/10/76            |
|   |                     | CA-A- 1062400              | 11/09/79            |
|   |                     | US-A- 4217428              | 12/08/80            |
|   |                     | US-A- 4072813              | 07/02/78            |
| US-A- 4299745                             | 10/11/81            | NONE                       |                     |



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